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date: April 27, 2017*to:* Distribution*from:* Kevin N. Long (1554) and Judith A. Brown (1554)*subject:* A Linear Viscoelastic Model Calibration of Sylgard® 184 (SAND2017-???? UUR)

Executive Summary

We calibrate a linear thermoviscoelastic model for solid Sylgard 184 (90-10 formulation), a lightly cross-linked, highly flexible isotropic elastomer for use both in Sierra / Solid Mechanics via the Universal Polymer Model as well as in Sierra / Structural Dynamics (Salinas) for use as an isotropic viscoelastic material. Material inputs for the calibration in both codes are provided. The frequency domain master curve of oscillatory shear was obtained from a report from Los Alamos National Laboratory (LANL) [4]. However, because the form of that data is different from the constitutive models in Sierra, we also present the mapping of the LANL data onto Sandia's constitutive models. Finally, blind predictions of cyclic tension and compression out to moderate strains of 40 and 20% respectively are compared with Sandia's legacy cure schedule material [2]. Although the strain rate of the data is unknown, the linear thermoviscoelastic model accurately predicts the experiments out to moderate strains for the slower strain rates, which is consistent with the expectation that quasi-static test procedures were likely followed. This good agreement comes despite the different cure schedules between the Sandia and LANL data.

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1 The Universal Polymer Model Theory Under Linear Viscoelastic Conditions

Our objective is to calibrate a linear thermo-viscoelastic model for Sylgard 184 (90 part by weight (PBW) component A to 10 PBW cross-linker component B) for use in Sierra / Solid Mechanics [6]. While there are several models that can represent linear viscoelastic materials, a subset including the Universal Polymer Model (UPM), Viscoelastic Swanson Model, and the HyperYeohDamage model, we seek to use the UPM model as it is the flexible and most production hardened model available viscoelastic materials at moderate to small deformations. Sylgard 184 is an elastomer, and large deformations are expected, but we do not at this time have data to calibrate the behavior in the huge (several hundred percent strain) regime. Consequently, the UPM model is a good choice.

Here we briefly reproduce the universal polymer model theory UPM specialized for linear thermo-viscoelasticity as laid out in materials manual for Lamé manual for materials in Sierra / Solid Mechanics [6]. Following the source work of the UPM model, which labeled it the Simplified Potential Energy Clock Model but here we will continue to call it by UPM as it is used in Sierra / Solid Mechanics reference [1], the strain measure is approximated from the integrated unrotated rate of deformation tensor, which we label $\boldsymbol{\epsilon}$,

$$\boldsymbol{\epsilon} = \int_0^\infty (\mathbf{R}^T \mathbf{D} \mathbf{R}) ds, \quad \mathbf{D} = \frac{1}{2} (\mathbf{L} + \mathbf{L}^T), \quad \mathbf{F} = \mathbf{R} \mathbf{U}. \quad (1)$$

Here, \mathbf{F} , \mathbf{R} , \mathbf{U} , \mathbf{L} , and \mathbf{D} are the deformation gradient, rotation, material stretch, velocity gradient, and rate of deformation tensors standard in Lagrangian continuum mechanics.

The UPM model allows the user to initiate an analysis from a stress-free temperature, T_{sf} , that is different from the reference temperature, T_{ref} , at which all material properties are defined. Here we briefly summarize the constitutive equations. The model is derived from a Helmholtz Free Energy, but we begin directly with the (unrotated) Cauchy Stress and refer the reader to reference [1] for more detail:

$$\begin{aligned} \boldsymbol{\sigma} = & (K_G[T] - K_\infty[T]) \int_0^t ds f_v [t' - s'] \frac{dI_1}{ds} \mathbf{1} \dots \\ & \dots - (K_G[T] \delta + G[T] - K_\infty[T] \delta_\infty[T]) \int_0^t ds f_v [t' - s'] \frac{dT}{ds} \mathbf{1} \dots \\ & \dots + 2(G_G[T] - G_\infty[T]) \int_0^t ds f_s [t' - s'] \frac{d(\text{dev} \boldsymbol{\epsilon})}{ds} ds \dots \\ & \dots + (K_\infty[T] I_1 - K_\infty[T] \delta_\infty[T] (T - T_{sf})) \mathbf{1} + 2G_\infty[T] \text{dev} \boldsymbol{\epsilon} \end{aligned} \quad (2)$$

The last collection of terms in 2 furnish the time-independent rubbery response. The variables in equation 2 are:

$$I_1 = \mathbf{1} : \boldsymbol{\epsilon} = \text{tr} \boldsymbol{\epsilon}, \quad \text{dev} \boldsymbol{\epsilon} = \boldsymbol{\epsilon} - I_1 \mathbf{1}, \quad (3)$$

$$G_G[T] = G_G^{\text{ref}} + \frac{dG_G}{dT}(T - T_{\text{ref}}), \quad G_{\infty}[T] = G_{\infty}^{\text{ref}} + \frac{dG_{\infty}}{dT}(T - T_{\text{ref}}), \quad (4)$$

$$K_G[T] = K_G^{\text{ref}} + \frac{dK_G}{dT}(T - T_{\text{ref}}), \quad K_{\infty}[T] = K_{\infty}^{\text{ref}} + \frac{dK_{\infty}}{dT}(T - T_{\text{ref}}), \quad (5)$$

$$\delta_G[T] = \alpha_G^{\text{ref}} + \frac{d\alpha_G}{dT}(T - T_{\text{ref}}), \quad \delta_{\infty}[T] = \alpha_{\infty}^{\text{ref}} + \frac{d\alpha_{\infty}}{dT}(T - T_{\text{ref}}), \quad (6)$$

The first three terms in Equation 2 represent the material's viscoelastic response to changes in volume strain, temperature, and shear deformation. Two relaxation functions are used to characterize the thermal/volumetric (f_v) and shear (f_s) relaxation responses. The model assumes the thermal and volumetric relaxation responses are identical. Otherwise, f_v and f_s are typically quite different and are expressed as a Prony series:

$$f_v[x] = \sum_{i=1}^N w_i \exp\left(-\frac{x}{\tau_i}\right), \quad f_s[x] = \sum_{j=1}^M w_j \exp\left(-\frac{x}{\tau_j}\right). \quad (7)$$

These relaxation functions describe the material's response to a suddenly applied volumetric/thermal or shear perturbation at the reference temperature where, under certain conditions, the material and laboratory time scales are equivalent. In Equation 2, the viscous terms (non-rubbery) involve hereditary integrals over the difference in material time from $s = 0$ to $s = t$, which is the current laboratory time. An increment in material time, dt' , and the laboratory time, dt , are related through the (highly) history dependent shift factor, a , such that the difference in material time, $t' - s'$, is related to the corresponding difference in laboratory time, $t - s$ through:

$$adt' = dt, \quad t' - s' = \int_{u=s}^{u=t} \frac{du}{a[u]}. \quad (8)$$

If the material time scale is very slow compared to the laboratory time, then $a \gg 1$, which is often the case inside and below the glass transition for typically glassy materials. For elastomers (materials well above the glass transition), the shift factor will not be much greater than 1.

The shift factor is instantaneously defined through:

$$\log_{10} a = \frac{-C_1 N}{C_2 + N}, \quad (9)$$

$$N[t] = T - T_{\text{ref}} - \int_0^t f_v[t' - s'] \frac{dT}{ds} ds + C_3 \left(I_1 - \int_0^t f_v[t' - s'] \frac{dI_1}{ds} ds \right) \quad (10)$$

$$\dots + C_4 \int_{u=0}^{u=t} \int_{s=0}^{s=t} \left(f_s[t' - s', t' - u'] \frac{d(\text{dev} \boldsymbol{\epsilon})}{ds} : \frac{d(\text{dev} \boldsymbol{\epsilon})}{du} ds du \right).$$

However, under *linear thermoviscoelastic* conditions, $C_3 = C_4 = 0$ such that only temperature and temperature history drives the shift factor evolution. However, we found that in large thermal cycles (still above the glass transition), the parameter, C_3 , was necessary to stabilize convergence of the model although it had little impact on the simulations.

The key physics in the model comes from Equation 9. Temperature rise (generally) causes N to increase, and hence the material shift factor shrinks (the material time scale speeds up). Shrinking the volume generally causes the shift factor to increase as if the temperature had been decreased (as long as $C_3 > 0$ otherwise there is no change of the clock with volume change). With $C_4 = 0$, there is no change in the shift factor with shear deformation.

It is desirable to relate a special case of the model to the Williams-Landel-Ferry (WLF) form because how time-temperature superposition fitting is typically performed [3]. To do, one can show that the parameters C_1 and C_2 relate to the WLF coefficients $\hat{C}_1 = C_1$ and $\hat{C}_2 = C_2 (1 + C_3 \alpha_\infty^{\text{ref}})$.

2 Master Curve Data from Los Alamos and How that Data is Mapped onto the Theory in Section 1

Workers at Los Alamos National Laboratory (LANL) performed a series of standard isothermal frequency sweeps in an oscillatory shear setup and analyzed with time temperature superposition (TTS) the in-phase (storage) and out-of-phase (loss) moduli vs. frequency. As is standard, they built a master curve of these quantities at 23 C for Sylgard 184 [4]. Since the form of their viscoelastic model theory is different than that used by the UPM model, the purpose of this section is to discuss how we mapped over their data and TTS master curve onto the UPM model for use in Sierra. Because the LANL report examines only the linear viscoelastic strain response, we only discuss the mapping of the second relaxation function in the UPM model (f_s). Small strain and stress measures are considered in both cases although the UPM model inherently involves the finite deformation strain(-like) measure of the integrated rate of (unrotated) deformation.

First we outline the UPM constitutive response under pure shear. The stress response is:

$$\sigma_{\text{UPM}} = 2(G_g - G_\infty) \int_0^t f_s(t' - s') \frac{d\gamma[s]}{ds} ds + G_\infty \gamma[s], \quad (11)$$

where G_g and G_∞ are the glassy and rubbery shear moduli at a particular temperature (not UPMified Eq. 11) and $\gamma[s]$ is the shear strain parameterized with time s . Here, $t' - s'$ represents the difference in material time evaluated through a time-temperature shift factor history as:

$$t' - s' = \int_s^t \frac{du}{a[u]}, \quad a[t]dt' = dt \quad (12)$$

Here, $a[u]$ represents the time shift factor between an increment in material time, dt' , and an increment in laboratory time, dt . For linear thermoviscoelasticity, the material time is an explicit function of the temperature. The LANL and UPM formulations both use a Williams-Landel-Ferry two parameter model that ultimately is used to fit master (relaxation function) curve data.

$$\log_{10} a = \frac{-\hat{C}_1(T - T_{\text{ref}})}{\hat{C}_2 + (T - T_{\text{ref}})}, \quad (13)$$

where C_1 and C_2 are constants used to shift data at different temperatures to form a master curve. The shear relaxation function, f_s , may always be expanded in a Prony series form:

$$f_s[x] = \sum_{i=1}^N f_i \exp\left(-\frac{x}{\tau_i}\right), \quad (14)$$

wherein f_i and τ_i are the i^{th} Prony weight and relaxation time constant.

Now, we outline the model used in the LANL report. The shear response is again:

$$\sigma_{\text{LANL}} = 2 \int_0^t G(t' - s') \frac{d\gamma[s]}{ds} ds, \quad (15)$$

where the shear relaxation function is:

$$G(t' - s') = G_\infty[T] \left(1 + \sum_{i=1}^N \chi_i \exp\left(-\frac{t' - s'}{\tau_i}\right) \right) \quad (16)$$

We want the two shear stress responses to be identical, and so we enforce:

$$\begin{aligned} \sigma_{\text{UPM}} - \sigma_{\text{LANL}} &= 0 \dots \\ \dots &= 2 \int_0^t \sum_i^N [(G_g - G_\infty)f_i - G_\infty\chi_i] \exp\left(-\frac{t' - s'}{\tau_i}\right) \frac{d\gamma[s]}{ds} ds. \end{aligned} \quad (17)$$

Therefore, we require that:

$$f_i = \frac{G_\infty\chi_i}{G_g - G_\infty}, \quad (18)$$

for each Prony term is satisfied. Note that Equation 18 is only rigorously true if G_g and G_∞ have identical temperature scalings. In general, that is not true for the UPM model, but here, we require it for this model to model mapping. All we have left to match is the glassy modulus, G_g . However, first, let us consider which parameters directly map over from the LANL formalism to the UPM model (ie. they are the same in both):

- The rubbery modulus, G_∞ , and the temperature dependence is straightforward to apply to the UPM model.
- The relaxation UPMtra, $\{\tau_i\}$, used to represent the master relaxation curve in the time domain at the reference temperature.
- WLF parameters and reference temperature of the master curve, \hat{C}_1 , \hat{C}_2 , and T_{ref} .

To identify the glassy modulus, G_g , consider the case when a strain event suddenly occurs and the material time scale is much slower than the laboratory time scale (such as if $T \ll T_{\text{ref}}$). In this regime, the arguments to the Prony series approach zero. Again, we equate the stress responses in this regime:

$$\begin{aligned} \sigma_{\text{UPM}} - \sigma_{\text{LANL}} = 0 &= G_\infty\gamma + (G_g - G_\infty)\gamma \sum_i^N f_i - G_\infty\gamma \left(1 + \sum_i^N \chi_i \right) \rightarrow \dots \\ \dots G_g &= G_\infty \left(1 + \sum_i^N \chi_i \right). \end{aligned} \quad (19)$$

Here, we have assumed that $\sum_i^N f_i = 1$, which may not be required in the UPM model but is standard in model parameterizations and generally recommended so that the above analysis defines the glassy response of a material. Given the relationship between G_g and the LANL parameters, the individual Prony weights in the UPM model can be further simplified (again assuming that $\sum_i^N f_i = 1$):

$$f_i = \frac{\chi_i}{\sum_{k=1}^N \chi_k}, \quad (20)$$

3 Sylgard 184 Calibration to the LANL Shear Master Curve Data and Comparison with Room Temperature Tension and Compression Experiments

Using time temperature superposition of oscillatory torsional Dynamic Mechanical Analysis (DMA) from -30 to 100C and frequencies 0.01 to 50 Hz in strain control mode, workers at Los Alamos National Laboratory (LANL) used time-temperature superposition ([3]) and determined the temperature dependent shift factor (Table 2 in [4]), a Prony series decomposition of shear relaxation function (Table 3 in [4]), and the equilibrium shear modulus vs. temperature (Table 4 in [4]). Using these data and the mapping method in section 2, a calibration for the 90-10 formulation of Sylgard 184 was determined for the UPM model. The bulk modulus (920 MPa) was taken for the legacy cure schedule from the Polymer website [2]. The WLF shift factor calibration is provided in Figure 1(a) while the equilibrium shear modulus evolution with temperature is given in Figure 1(b). We note that the parameter $C_3 = 1000 K^{-1}$, which mimics the volume strain dependence of the shift factor in glassy epoxies and has little effect on the shift factor well above the glass transition (about -100 C [2]). Simplified thermal expansion data was taken from the polymer website as well, and we recommend that the stress-free temperature should either be the curing temperature (either room temperature or 71 C) [2]. The calibrated model for Sierra/Solid Mechanics (see [5]) is:

```
##-----
## Sylgard_184_90_10 LANL and Legacy Schedule from the Polymer Website
## SI UNITS
##-----
begin Material Sygard184_ShearViscoLANL
    DENSITY          = 1003 # kg / m^3
    bulk modulus      = 0.92E9 # Pa
    shear modulus      = 0.61E6 # Pa

    ###---Universal Polymer model-----
    ### reduced here to an LVE model by setting clock c3 and clock c4 = 0
begin parameters for model universal_polymer
    ## The following are not used but may be required to run the model
    wwbeta 1 = 0.14
    wwtau 1 = 6 ## s
    wwbeta 2 = 0.0
    wwtau 2 = 0.0 ## s
```

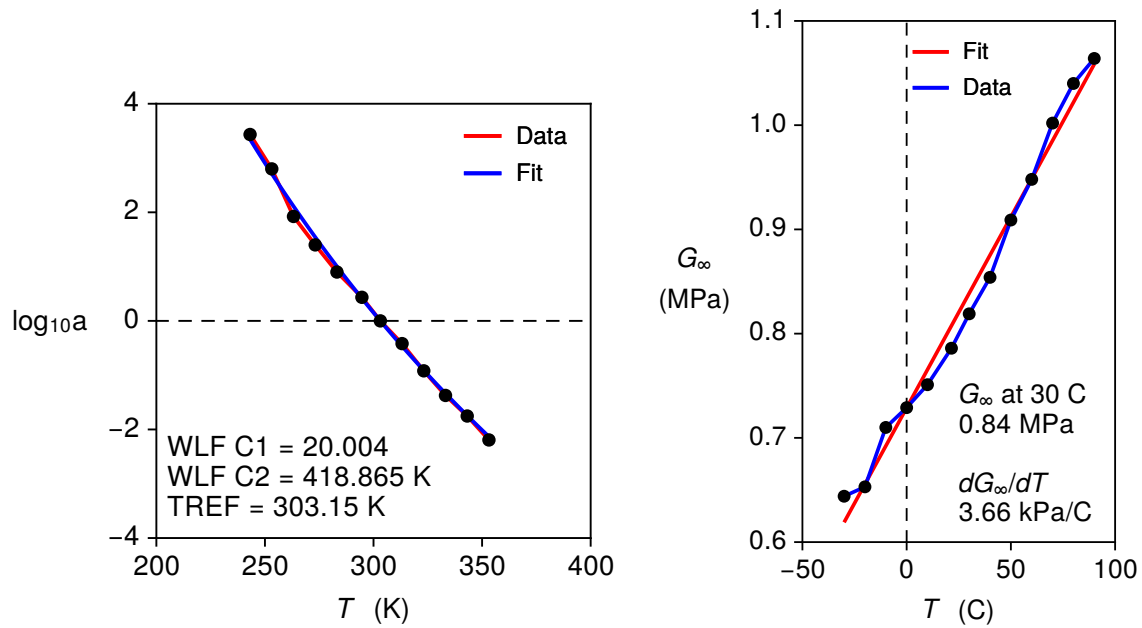


Figure 1. Equilibrium shear modulus vs. Temperature and the WLF calibration to the shift factor vs. temperature (all data is from [4])

```

UPMtrum start time = 0.0
UPMtrum end time = 0.0
log time increment = 0.0
## Resume used parameters
## By making the bulk glassy and rubbery moduli the same,
## there is no volumetric viscoelasticity
bulk glassy 0 = 0.92E9      ## Pa
bulk rubbery 0 = 0.92E9    ## Pa
volcte glassy 0 = 0.00017  ## 1/K
volcte rubbery 0 = 0.0006  ## 1/K
### Begin LANL fits:
reference temperature = 303.15 # K
Shear Glassy 0 = 3.621783e+06 # Pa
Shear Glassy 1 = 3.662777e+03 # Pa per degree K
Shear Rubbery 0 = 8.387312e+05 # Pa
Shear Rubbery 1 = 3.662777e+03 # Pa per degree K
WLF C1 = 20.003660
WLF C2 = 418.862744 # K
clock c1 = 0
clock c2 = 0
clock c3 = 1000 # 1/K
clock c4 = 0

```

```
clock c5 = 0
clock c6 = 0
filler vol fraction = 0.0
stress free temperature = {TCURE}  ## Should be about Room Temperature (20 C) c
### Direct Prony Series representation of the shear spectrum
relax time 1 = 1.00000e-06 # Units of time (seconds)
relax time 2 = 3.16000e-06
relax time 3 = 1.00000e-05
relax time 4 = 3.16000e-05
relax time 5 = 1.00000e-04
relax time 6 = 3.16000e-04
relax time 7 = 1.00000e-03
relax time 8 = 3.16000e-03
relax time 9 = 1.00000e-02
relax time 10 = 3.16000e-02
relax time 11 = 1.00000e-01
relax time 12 = 3.16000e-01
relax time 13 = 1.00000e+00
relax time 14 = 3.16000e+00
relax time 15 = 1.00000e+01
relax time 16 = 3.16000e+01
relax time 17 = 1.00000e+02
relax time 18 = 3.16000e+02
relax time 19 = 1.00000e+03
relax time 20 = 3.16000e+03
f2 1 = 5.06098e-01
f2 2 = 0.00000e+00
f2 3 = 1.27611e-01
f2 4 = 7.55463e-02
f2 5 = 6.61488e-02
f2 6 = 5.68130e-02
f2 7 = 4.20298e-02
f2 8 = 3.56497e-02
f2 9 = 2.57836e-02
f2 10 = 1.89884e-02
f2 11 = 1.26279e-02
f2 12 = 1.09535e-02
f2 13 = 3.99764e-03
f2 14 = 6.17595e-03
f2 15 = 2.26342e-03
f2 16 = 2.91730e-03
f2 17 = 2.04165e-03
f2 18 = 1.72507e-03
f2 19 = 8.52181e-04
```



```
f2 20 = 1.77735e-03
# NOT USED but required volumetric Prony series
f1 1 = 5.06098e-01
f1 2 = 0.00000e+00
f1 3 = 1.27611e-01
f1 4 = 7.55463e-02
f1 5 = 6.61488e-02
f1 6 = 5.68130e-02
f1 7 = 4.20298e-02
f1 8 = 3.56497e-02
f1 9 = 2.57836e-02
f1 10 = 1.89884e-02
f1 11 = 1.26279e-02
f1 12 = 1.09535e-02
f1 13 = 3.99764e-03
f1 14 = 6.17595e-03
f1 15 = 2.26342e-03
f1 16 = 2.91730e-03
f1 17 = 2.04165e-03
f1 18 = 1.72507e-03
f1 19 = 8.52181e-04
f1 20 = 1.77735e-03
end parameters for model universal_polymer
```

With the model calibrated, we compare to moderate deformation tension and compression experimental data from the polymer website [2]. Because the strain rate of the experimental data is unknown, we simulate three different strain rates to bound the experimental behavior. A single finite element under homogenous motion is deformed in these two uniaxial stress boundary value problems under displacement control. Each element is loaded at a fixed engineering strain rate to a target strain, held for 0.1 seconds to allow for stress relaxation, and then unloaded back to zero displacement. At the faster strain rates, a hysteresis is observed that arises solely from viscous dissipation, which grows considerably at the faster strain rate. Compared with the data, we conjecture the experiments were performed at reasonably slow strain rates (though still some viscous dissipation is evident). The tension and compression comparisons are shown in Figure 2. No effort was made to tweak the equilibrium shear modulus with shear deformation to better match the finite strain results although that can be done in future efforts.

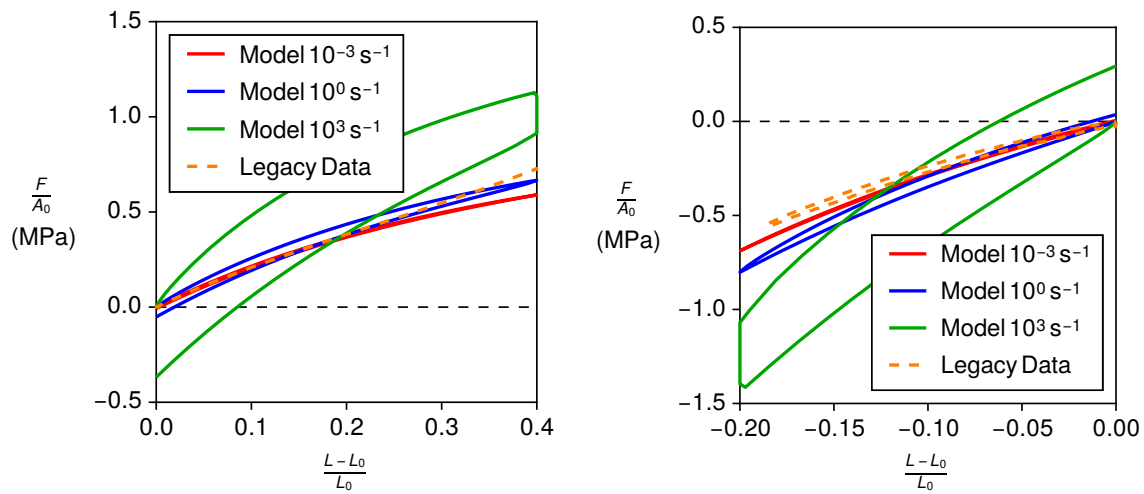


Figure 2. Tension and compression comparison with legacy data from [2] performed at 23 C. The strain rate of the data is unknown, so three characteristic (slow, moderate, and fast) strain rates are simulated in part to bound the data. A 0.1 second hold is included in both simulations followed by an unload step at the same engineering strain rate (as the loading step). Experimentally, no hold exists, and the tensile data only involves an initial load step (no unload). It appears that the data was performed at a relatively slow strain rate of less than 1 per second. Significant hysteresis (dissipation) is observed at the faster strain rate in both cases.

Finally, the parameterization for the model suitable for linear viscoelasticity in Sierra / Structural Dynamics (Salinas) in time-domain simulations is provided:

```
#####----- Sierra SD (Salinas) Input (SI UNITS) -----#####
MATERIAL 4 # Not sure about number 4
ISOTROPIC VISCOELASTIC

## NOTE, this is approximate for SOLID Sylgard 184 (90/10 mixture)
DENSITY      1003      # kg / m^3.

T_0           303.15    # Kelvin
T_CURRENT     298.00    # Kelvin.  CHANGE THIS VALUE based on your analysis

C_1           20.003669 # None. First WLF Coefficient
C_2           418.862744 # Kelvin. Second WLF Coefficient

aT_1          1.0 # NONE? coef. to modify the WLF below the ref temperature (?)
aT_2          1.0 # NONE? "below or above?"

K_g           0.92E9    # Pa, Glassy bulk modulus
K_inf         0.92E9    # PA, Rubbery bulk modulus
### Formulations between SM and SD are a bit different here. Hence the minus
G_g           {3.621783e+06-8.387312e+05} #Pa, Glassy shear modulus.
G_inf         8.387312e+05 #Pa, Rubbery shear modulus

K_relax
  T(1)=1.00000e-06
  T(2)=3.16000e-06
  T(3)=1.00000e-05
  T(4)=3.16000e-05
  T(5)=1.00000e-04
  T(6)=3.16000e-04
  T(7)=1.00000e-03
  T(8)=3.16000e-03
  T(9)=1.00000e-02
  T(10)=3.16000e-02
  T(11)=1.00000e-01
  T(12)=3.16000e-01
  T(13)=1.00000e+00
  T(14)=3.16000e+00
  T(15)=1.00000e+01
  T(16)=3.16000e+01
  T(17)=1.00000e+02
  T(18)=3.16000e+02
```

T(19)=1.00000e+03

T(20)=3.16000e+03

K_coeff

M(1)=5.06098e-01

M(2)=0.00000e+00

M(3)=1.27611e-01

M(4)=7.55463e-02

M(5)=6.61488e-02

M(6)=5.68130e-02

M(7)=4.20298e-02

M(8)=3.56497e-02

M(9)=2.57836e-02

M(10)=1.89884e-02

M(11)=1.26279e-02

M(12)=1.09535e-02

M(13)=3.99764e-03

M(14)=6.17595e-03

M(15)=2.26342e-03

M(16)=2.91730e-03

M(17)=2.04165e-03

M(18)=1.72507e-03

M(19)=8.52181e-04

M(20)=1.77735e-03

G_relax

T(1)=1.00000e-06

T(2)=3.16000e-06

T(3)=1.00000e-05

T(4)=3.16000e-05

T(5)=1.00000e-04

T(6)=3.16000e-04

T(7)=1.00000e-03

T(8)=3.16000e-03

T(9)=1.00000e-02

T(10)=3.16000e-02

T(11)=1.00000e-01

T(12)=3.16000e-01

T(13)=1.00000e+00

T(14)=3.16000e+00

T(15)=1.00000e+01

T(16)=3.16000e+01

T(17)=1.00000e+02

T(18)=3.16000e+02

T(19)=1.00000e+03

T(20)=3.16000e+03

```
T(21)=1.00000e+04
T(22)=3.16000e+04
G_coeff
M(1)=5.06098e-01
M(2)=0.00000e+00
M(3)=1.27611e-01
M(4)=7.55463e-02
M(5)=6.61488e-02
M(6)=5.68130e-02
M(7)=4.20298e-02
M(8)=3.56497e-02
M(9)=2.57836e-02
M(10)=1.89884e-02
M(11)=1.26279e-02
M(12)=1.09535e-02
M(13)=3.99764e-03
M(14)=6.17595e-03
M(15)=2.26342e-03
M(16)=2.91730e-03
M(17)=2.04165e-03
M(18)=1.72507e-03
M(19)=8.52181e-04
M(20)=1.77735e-03
```

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